

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

979/50806

U.S. APPLICATION NO. (if known, see
37 CFR 1.52)

INTERNATIONAL APPLICATION NO.

PCT/EP00/05377

INTERNATIONAL FILING DATE

10 June 2000

PRIORITY DATE CLAIMED

29 June 1999

TITLE OF INVENTION

GASTIGHT PRISMATIC NICKEL-METAL HYDRIDE CELL

APPLICANT(S) FOR DO/EO/US

Gabor BENCZUR-UER-MOESSY

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

- | | | |
|-----|-------------------------------------|---|
| 1. | <input checked="" type="checkbox"/> | This is a FIRST submission of items concerning a filing under 35 U.S.C. 371 |
| 2. | <input type="checkbox"/> | This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371 |
| 3. | <input checked="" type="checkbox"/> | This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay Examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). |
| 4. | <input checked="" type="checkbox"/> | A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. |
| 5. | <input checked="" type="checkbox"/> | A copy of the International Application as filed (35 U.S.C. 371(c)(2)). |
| | <input type="checkbox"/> | a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). |
| | <input type="checkbox"/> | b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau (PCT/IB/308) |
| | <input type="checkbox"/> | c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US) |
| 6. | <input checked="" type="checkbox"/> | A translation of the International Application into English (35 U.S.C. 371(c)(2)). |
| 7. | <input type="checkbox"/> | Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) |
| | <input type="checkbox"/> | a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). |
| | <input type="checkbox"/> | b. <input type="checkbox"/> have been transmitted by the International Bureau. |
| | <input type="checkbox"/> | c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. |
| | <input type="checkbox"/> | d. <input type="checkbox"/> have not been made and will not be made. |
| 8. | <input type="checkbox"/> | A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). |
| 9. | <input checked="" type="checkbox"/> | An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)) (Unexecuted, pages 1-2) |
| 10. | <input type="checkbox"/> | A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). |

Item 11. to 16. below concern other document(s) or information included:

- | | | |
|-----|-------------------------------------|---|
| 11. | <input checked="" type="checkbox"/> | An Information Disclosure Statement under 37 CFR 1.97 and 1.98 |
| 12. | <input type="checkbox"/> | An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. |
| 13. | <input checked="" type="checkbox"/> | A FIRST preliminary amendment. |
| | <input type="checkbox"/> | A SECOND or SUBSEQUENT preliminary amendment. |
| 14. | <input checked="" type="checkbox"/> | A substitute specification and marked-up copy thereof. |
| 15. | <input type="checkbox"/> | A change of power of attorney and/or address letter. |
| 16. | <input type="checkbox"/> | Other items or information: |
| | a. | 1 Sheet of Drawings, showing Figure 1. |
| | b. | International Preliminary Examination Report and its Transmittal |
| | c. | International Search Report w/references |

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)		INTERNATIONAL APPLICATION NO.		ATTORNEYS DOCKET NUMBER	
10/019488		PCT/EP00/05377		979/50806	
17. <input checked="" type="checkbox"/> The following fees are submitted:				CALCULATIONS	PTO USE ONLY
Basic National Fee (37 CFR 1.492(a)(1)-(5)):					
Search Report has been prepared by the EPO or JPO \$ 890.00				\$890.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482)				\$ 690.00	
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) \$ 740.00					
Neither international preliminary examination fee (37 CFR 1.482) nor International search fee (37 CFR 1.445(a)(2)) paid to USPTO \$ 1000.00					
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$ 100.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than [] 20 [X] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$130.00	
Claims	Number Filed	Number Extra	Rate		
Total Claims	16 - 20 =	0	X \$18.00	\$	
Independent Claims	1 - 3 =	0	X \$84.00	\$	
Multiple dependent claims(s) (if applicable)			+ \$280.00	\$	
TOTAL OF ABOVE CALCULATIONS=				\$1,020.00	
Applicant claims Small Entity Status (See 37 CFR §1.27) [] yes [X] no. Reduction by 1/2 for filing by small entity, if applicable.				\$	
SUBTOTAL =				\$1,020.00	
Processing fee of \$130.00 for furnishing the English translation later than [] 20 [X] 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$1,020.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28,3.31). \$40.00 per property +				\$	
TOTAL FEE ENCLOSED =				\$1,020.00	
				Amount to be:	
				refunded \$	
				Charged \$	
<p>a. <input checked="" type="checkbox"/> A check in the amount of \$1,020.00 for the filing fee is enclosed</p> <p>b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees, which may be required, or credit any overpayment to Deposit Account No. 05-1323. (Attorney Docket #979/50608). A duplicate copy of this sheet is enclosed.</p> <p>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</p>					
<p>SEND ALL CORRESPONDENCE TO:</p> <p>Crowell & Moring, L.L.P.</p> <p>Intellectual Property Group</p> <p>P.O. Box 14300</p> <p>Washington, D.C. 20044-4300</p> <p>Tel. No. (202) 624-2500</p> <p>Fax No. (202) 628-8844</p>					
				<p><i>Warren A. Zitlau</i></p> <p>SIGNATURE</p> <p>Warren A. Zitlau</p> <p>NAME</p> <p>39,085</p> <p>REGISTRATION NUMBER</p> <p>December 31, 2001</p> <p>DATE</p>	

Attorney Docket: 979/50806
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: GABOR BENCZUR-UERMOESSY
Serial No.: TO BE ASSIGNED PCT No.: PCT/EP00/05377
Filed: DECEMBER 31, 2001
Title: GASTIGHT PRISMATIC NICKEL-METAL HYDRIDE CELL

PRELIMINARY AMENDMENT

Box PCT
Commissioner for Patents
Washington, D.C. 20231

DECEMBER 31, 2001

Sir:

Please enter the following amendments to the claims and abstract prior to the examination of the application.

IN THE CLAIMS:

Please cancel claims 1-13.

Please add the following new claims:

14. (New) A gastight cell for the storage of electrochemical energy, comprising at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and an alkaline electrolyte or an alkaline electrolyte mixture, wherein one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.

15. (New) The cell as claimed in claim 14, comprising an electrode assembly comprising n positive electrodes and $(n+1)$ negative electrodes, with two outmost negative electrodes, which are flanked by in each case one transport element.

16. (New) The cell as claimed in claim 14, wherein one or more negative electrodes are split into two parts, the two parts being separated from one another by a hydrophobic, gas-permeable transport element.

17. (New) The cell as claimed in claim 16, comprising a plurality of positive and negative electrodes that are alternately arranged, wherein every second negative electrode is split into two parts.

18. (New) The cell as claimed in one of claim 16, wherein the two parts of the split negative electrodes each has half the thickness or half the capacitance of an unsplit negative electrode.

19. (New) The cell as claimed in claim 14, wherein the transport element is a hydrophobic nonwoven layer.

20. (New) The cell as claimed in claim 19, wherein the hydrophobic nonwoven layer comprises electrolyte-repelling polypropylene fibers

21. (New) The cell as claimed in claim 14, wherein the positive electrodes are fibrous-structure framework electrodes.

22. (New) The cell as claimed in claim 14, wherein the separator comprises nonwoven polyamide fiber or hydrophilic nonwoven polypropylene fiber.

23. (New) The cell as claimed in claim 14, wherein the negative electrode comprises a metallic substrate material, to which an active compound is

applied, the active compound being obtainable from a paste which comprises a dry fraction and a liquid fraction, the dry fraction comprising a mixture of a pulverulent storage alloy for hydrogen, soot and polytetrafluoroethylene (PTFE), and the liquid fraction comprising a mixture of water and an alcohol which has 3 to 6 C atoms.

24. (New) The cell as claimed in claim 23, wherein the dry fraction comprising particles of the storage alloy being covered with polytetrafluoroethylene in the manner of fibrils.

25. (New) The cell as claimed in claim 14, wherein the dry fraction comprises 85 to 95 parts of the alloy for storing hydrogen, 2 to 10 parts of soot and 3 to 8 parts of PTFE.

26. (New) The cell as claimed in claim 23, wherein the liquid fraction contains 30 to 70 parts by volume of water and 70 to 30 parts by volume of the alcohol.

27. (New) The cell as claimed in claim 23, wherein the dry fraction further comprises 0.2% by weight of polyethylene glycol.

28. (New) The cell as claimed in claim 23, wherein the liquid fraction comprises polyethylene glycol.

29. (New) The cell as claimed in claim 23, wherein the dry fraction and the liquid fraction has a mass ratio of 4:1 to 6:1.

IN THE ABSTRACT:

Please substitute the new Abstract of the Disclosure submitted herewith on a separate page for the original Abstract presently in the application.

REMARKS

Entry of the amendments to the specification, claims and abstract before examination of the application is respectfully requested. These claims have been amended to remove multiple dependencies and otherwise conform with U.S. practices, such that these claims platenably define over the art of record.

If there are any questions regarding this Preliminary Amendment or this application in general, a telephone call to the undersigned would be appreciated since this should expedite the prosecution of the application for all concerned.

Respectfully submitted,



Warren A. Zitlau
Registration No. 39,085
Kening Li
Registration No. 44,872

CROWELL & MORING, LLP
Intellectual Property Group
P.O. Box 14300
Washington, DC 20044-4300
Telephone No.: (202) 624-2500
Facsimile No.: (202) 628-8844
WAZ:KL:tlm
(CAM #: 80449.046)

ABSTRACT OF THE DISCLOSURE

A gastight nickel/metal hydride cell for the storage of electrochemical energy, having at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and an alkaline electrolyte or an alkaline electrolyte mixture, in that one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.

**VERSION WITH MARKINGS TO SHOW CHANGES MADE TO THE
ABSTRACT**

ABSTRACT OF THE DISCLOSURE

[The present invention relates to a] A gastight nickel/metal hydride cell for the [electrochemical] storage of electrochemical energy, having at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and [having] an alkaline electrolyte or an alkaline electrolyte mixture, in that one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.

Attorney Docket: 979/50806
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: GABOR BENCZUR-UERMOESSY
Serial No.: TO BE ASSIGNED PCT NO.: PCT/EP00/05377
Filed: DECEMBER 31, 2001
Title: GASTIGHT PRISMATIC NICKEL-METAL HYDRIDE CELL

SUBMISSION OF SUBSTITUTE SPECIFICATION

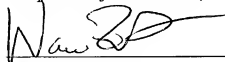
Assistant Commissioner for Patents
Washington, D.C. 20231

December 31, 2001

Sir:

Attached is a Substitute Specification and a marked-up copy of the original specification. I certify that said substitute specification contains no new matter and includes the changes indicated in the marked-up copy of the original specification.

Respectfully submitted,



Warren A. Zitlau
Registration No. 39,085
Kening Li, Ph.D.
Registration No. 44,872

CROWELL & MORING, LLP
P.O. Box 14300
Washington, DC 20044-4300
Telephone No.: (202) 624-2500
Facsimile No.: (202) 628-8844
WAS:KL:tlm
(CAM #: 80449.046)

GASTIGHT, PRISMATIC NICKEL/METAL HYDRIDE CELL

BACKGROUND AND SUMMARY OF THE INVENTION

[0001] This is a national stage application of International Patent Application No. PCT/EP00/05377 filed on June 10, 2000 designating the United States of America, the entire disclosure of which is expressly incorporated by reference herein. Priority is claimed based on Federal Republic of Germany patent application No. 199 29 947.1, filed June 29, 1999.

[0002] The invention relates to a gastight cell for the storage of electrochemical energy, having at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and having an alkaline electrolyte or an alkaline electrolyte mixture.

[0003] Storage batteries for storing electrical energy in the form of chemical energy, which can then be removed again as electrical energy, have been known since the end of the nineteenth century. Even today, the lead storage battery is still in widespread use. In such a battery, the electrodes or plates comprise the active material, which is the actual energy store, and a lead support (grid), which holds the active material. There are also batteries with alkaline aqueous electrolytes.

[0004] All these galvanic elements substantially comprise the energy-storing electrodes of positive and negative polarity, the electrolyte, the separator between the electrodes, the cell or battery vessel and the current-carrying, connective inactive parts, such as the supply and discharge lines for the current to and from the electrodes. These also include substrate material, current discharge lugs, poles, pole bridges, pole screws, washers and pole connectors.

[0005] In a gastight nickel/metal hydride cell, the negative and positive electrodes are arranged alternately next to one another, for example in a prismatic or cuboidal housing, and are separated from one another by a separator. On account of the

normally absent freely mobile excess of electrolyte, each of the electrodes is in contact with the cell atmosphere, i.e. the gas space of the cell. The gases which are evolved during charging of the cell pass into this gas space and, in quiescent phases of the cell, react at the negative electrodes. For this purpose, the gases have to diffuse into the electrodes. By way of example, hydrogen is reincorporated in the lattice of the storage alloy of the negative electrode, until an equilibrium state is reached.

[0006] Cells of the generic type are described in EP 0 460 424 B1, EP 0 460 425 B1 and DE 39 29 306 C2.

[0007] A problem of these cells is that in quiescent phases charge balancing in all the negative electrodes of the cell is not possible, and an excess pressure of hydrogen and oxygen continues to obtain. In fact, auxiliary electrodes or special multilayer electrodes are required for charge balancing.

[0008] Therefore, the object of the present invention is to provide a cell of the abovementioned type in which charge balancing is possible in the negative electrodes and the excess pressure is reduced with the minimum possible outlay.

[0009] The instant invention provides a cell comprising at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and an alkaline electrolyte or an alkaline electrolyte mixture, wherein one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.

[0010] Therefore, according to the invention, one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.

[0011] The gases of the cell atmosphere, namely hydrogen and oxygen, fill these transport elements and reach the pores in the negative electrodes, which are only

partially filled with electrolyte. There, the oxygen is quickly reduced or reacts with the hydrogen, which is present in excess, to form water. The gaseous hydrogen reacts with the storing alloy until thermodynamic equilibrium is reached. In this way, charge balancing is achieved in all the negative electrodes of the cell in quiescent phases, and the excess gas pressure is reduced. It is even possible to reverse the polarity of the cell, since the hydrogen which now evolves at the positive electrode reaches the transport elements and therefore the negative electrodes via the gas phase, and is then oxidized. This sequence of events can be detected by means of a discharge current, e.g. approximately -0.2 V cell voltage, which flows for an unlimited time.

[0012] Some advantageous refinements and preferred embodiment of the invention are described below. In the case of cells with high load-bearing capacities and electrode thicknesses of less than 0.5 mm, the outer negative electrodes are preferably flanked by a transport element. Consequently, the balancing operations described above take place relatively slowly.

[0013] When using cells with a thicker electrode, it is advantageous if one or more negative electrodes are split into two parts, the two parts being separated from one another by a hydrophobic, gas-permeable transport element. As a result, the balancing operations which have been described above take place relatively quickly.

[0014] Preferably, in the sequence of a plurality of positive and negative electrodes, every second negative electrode is split into two parts. The two parts of the split negative electrodes advantageously have half the thickness or half the capacitance of the unsplit negative electrodes.

[0015] The transport element is, for example, a hydrophobic nonwoven layer, preferably comprising electrolyte-repelling polypropylene fibers.

[0016] The positive electrodes are, for example, nickel oxide electrodes, preferably fibrous-structure framework electrodes, while the negative electrodes are hydrogen-

storing electrodes. The separators preferably comprise polyamide fiber nonwoven or hydrophilic polypropylene fiber nonwoven.

[0017] Other objects, advantages and novel features of the present invention will become apparent from the following detailed description of the invention when considered in conjunction with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

[0018] The sole figure shows a cell according to the present invention.

DETAILED DESCRIPTION OF THE DRAWINGS

[0019] A preferred embodiment of the cell according to the invention is diagrammatically depicted in Figure 1. The cell 1 has a prismatic housing 2 with a positive pole 3 and a negative pole 4. In the housing 2 there are positive nickel oxide electrodes 5 and negative electrodes 6a, 6b comprising a hydrogen-storage alloy, which are in each case separated from one another by a separator 7. All the electrodes are in communication with the gas space 8. Every second negative electrode 6b comprises two half part electrodes 10, 11 which are separated from one another by a gas-permeable transport element 12 in the form of a hydrophobic nonwoven layer.

[0020] The negative electrode is preferably equipped with a specifically set hydrophobic/hydrophilic balance. For this purpose, the active compound is obtainable from a paste which is composed of a dry fraction and a liquid fraction. In addition to a hydrogen-storage alloy and polytetrafluoroethylene (PTFE), the dry fraction also contains soot; the liquid fraction contains water and an alcohol with 3-6 C atoms, the particles of the storage alloy being covered with PTFE in the manner of fibrils.

[0021] The addition of soot is important to make the mixture easier to process. The addition of soot makes the mixture pasty and able to flow. In the electrode, the soot

promotes the electrical contact on a microscale (up to approximately 500 μm), i.e. it spans the distances and provides electrical contact between the openings or pores in the substrate material. These distances or openings, with a size of up to 500 μm , cannot be bridged by the particles in the paste or the active compound which can be obtained therefrom, since they generally have a diameter of only approximately 10 to 100 μm . Furthermore, the soot serves as an oxygen getter for protecting the oxygen-sensitive storage alloy. The PTFE is responsible for the hydrophobic properties of the electrode and enables the three-phase boundary to be set. The reduction of the oxygen and the release and uptake of the hydrogen in the working cell take place in the only partially wetted pores. The PTFE is also responsible for enabling the paste to flow and hold together in the mixing or shaping process. The alcohol is in turn responsible for the temporary wetting of the PTFE powder, since otherwise there would be no distribution through fibrillation in the mixing process.

[0022] The method for producing the electrode according to the invention is very simple. The components are mixed in a mixer until a cohesive paste is formed. The paste is shaped and combined with the metallic substrate material (for example expanded metal, fabric, grid, perforated sheet) of the electrode. This is an extraordinarily simple process sequence.

[0023] The electrode according to the invention means that it is also no longer necessary to use a PTFE dispersion with a high wetting agent content, which has to be removed by decomposition at elevated temperature (300°C), damaging the storage alloy, in conventional plastic-bonded storage electrodes.

[0024] The dry fraction contains 85-95 parts of the storage alloy, approximately 2-10 parts of soot and 3-8 parts of PTFE. The liquid fraction contains 30-70 parts by volume of water and approximately 70-30 parts by volume of the alcohol. Alcohols with a boiling point of the order of magnitude of approximately 100°C, i.e. for example n-butanol or n-propanol, are particularly suitable.

[0025] Furthermore, polyethylene glycol (PEG) may be included in the liquid fraction. The PTFE component means that the finished electrode can only be wetted by lye with extreme difficulty. Therefore, to achieve a sufficient uptake of electrolyte, a polyethylene glycol can be supplied with the make-up water. The proportion of polyethylene glycol is 0.05-0.2% (based on the dry fraction). It is preferable to use a polyethylene glycol with a molecular weight of between 10^5 and 5×10^6 g/mol. The alcohol used is preferably n-propanol or n-butanol. Depending on the soot content, the ratio of the dry fraction to the liquid fraction is between 4:1 and 6:1, based on mass.

[0026] The electrode according to the invention is preferably used in an alkaline storage battery with positive nickel oxide electrode.

[0027] The electrode according to the invention is produced by rolling a dough-like paste onto a structured metal substrate, such as for example an expanded metal or grid mesh. The dough-like paste is prefabricated in a first production step by a mixing and kneading process. The solid and liquid components are mixed in a kneading machine until a cohesive paste is formed, for example in a stable domestic kneading machine. The PTFE particles are fibrillated by the hard compound grains and hold the paste together. The electrode is shaped either by manual rolling or in a rolling train. Either a sheet is produced and is combined with the substrate after drying or the kneaded compound is applied directly to the substrate and is then dried.

[0028] The foregoing disclosure has been set forth merely to illustrate the invention and is not intended to be limiting. Since modifications of the disclosed embodiments incorporating the spirit and substance of the invention may occur to persons skilled in the art, the invention should be construed to include everything within the scope of the appended claims and equivalents thereof.

1/ptb

Gastight, prismatic nickel/metal hydride cell

The invention relates to a gastight cell for the storage of electrochemical energy, having at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and having an alkaline electrolyte or an alkaline electrolyte mixture.

(Storage) batteries for storing electrical energy in the form of chemical energy, which can then be removed again as electrical energy, have been known since the end of the last century. Even today, the lead storage battery is still in widespread use. In such a battery, the electrodes or plates comprise the active material, which is the actual energy store, and a lead support (grid), which holds the active material. There are also batteries with alkaline aqueous electrolytes.

All these galvanic elements substantially comprise the energy-storing electrodes of positive and negative polarity, the electrolyte, the separator between the electrodes, the cell or battery vessel and the current-carrying, connective inactive parts, such as the supply and discharge lines for the current to and from the electrodes. These also include substrate material, current discharge lugs, poles, pole bridges, pole screws, washers and pole connectors.

In a gastight nickel/metal hydride cell, the negative and positive electrodes are arranged alternately next to one another, for example in a prismatic or cuboidal housing, and are separated from one another by a separator. On account of the normally absent freely mobile excess of electrolyte, each of the electrodes is in contact with the cell atmosphere, i.e. the gas space of the cell. The gases which are evolved during charging of the cell pass into this gas space and, in quiescent phases of

the cell, react at the negative electrodes. For this purpose, the gases have to diffuse into the electrodes. By way of example, hydrogen is reincorporated in the lattice of the storage alloy of the negative electrode, until an equilibrium state is reached.

Cells of the generic type are described in EP 0 460 424 B1, EP 0 460 425 B1 and DE 39 29 306 C2.

A problem of these cells is that in quiescent phases charge balancing in all the negative electrodes of the cell is not possible, and an excess pressure of hydrogen and oxygen continues to obtain. In fact, auxiliary electrodes or special multilayer electrodes are required for charge balancing.

Therefore, the object of the present invention is to provide a cell of the abovementioned type in which charge balancing is possible in the negative electrodes and the excess pressure is reduced with the minimum possible outlay.

The solution consists in a cell having the features of claim 1.

Therefore, according to the invention, one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.

The gases of the cell atmosphere, namely hydrogen and oxygen, fill these transport elements and reach the pores in the negative electrodes, which are only partially filled with electrolyte. There, the oxygen is quickly reduced or reacts with the hydrogen, which is present in excess, to form water. The gaseous hydrogen reacts with the storing alloy until thermodynamic equilibrium is reached. In this way, charge balancing is achieved in all the negative electrodes of the cell in quiescent phases, and the excess gas pressure is reduced. It is even possible to reverse

the polarity of the cell, since the hydrogen which now evolves at the positive electrode reaches the transport elements and therefore the negative electrodes via the gas phase, and is then oxidized. This sequence of events can be detected by means of a discharge current, e.g. approximately -0.2 V cell voltage, which flows for an unlimited time.

Advantageous refinements will emerge from the subclaims. In the case of cells with high load-bearing capacities and electrode thicknesses of less than 0.5 mm, the outer negative electrodes are preferably flanked by a transport element. Consequently, the balancing operations described above take place relatively slowly.

When using cells with a thicker electrode, it is advantageous if one or more negative electrodes are split into two parts, the two parts being separated from one another by a hydrophobic, gas-permeable transport element. As a result, the balancing operations which have been described above take place relatively quickly.

Preferably, in the sequence of a plurality of positive and negative electrodes, every second negative electrode is split into two parts. The two parts of the split negative electrodes advantageously have half the thickness or half the capacitance of the unsplit negative electrodes.

The transport element is, for example, a hydrophobic nonwoven layer, preferably comprising electrolyte-repelling polypropylene fibers.

The positive electrodes are, for example, nickel oxide electrodes, preferably fibrous-structure framework electrodes, while the negative electrodes are hydrogen-storing electrodes.

The separators preferably comprise polyamide fiber nonwoven or hydrophilic polypropylene fiber nonwoven.

A preferred embodiment of the cell according to the invention is diagrammatically depicted in Figure 1. The cell 1 has a prismatic housing 2 with a positive pole 3 and a negative pole 4. In the housing 2 there are positive nickel oxide electrodes 5 and negative electrodes 6a, 6b comprising a hydrogen-storage alloy, which are in each case separated from one another by a separator 7. All the electrodes are in communication with the gas space 8. Every second negative electrode 6b comprises two half part electrodes 10, 11 which are separated from one another by a gas-permeable transport element 12 in the form of a hydrophobic nonwoven layer.

The negative electrode is preferably equipped with a specifically set hydrophobic/hydrophilic balance. For this purpose, the active compound is obtainable from a paste which is composed of a dry fraction and a liquid fraction. In addition to a hydrogen-storage alloy and polytetrafluoroethylene (PTFE), the dry fraction also contains soot; the liquid fraction contains water and an alcohol with 3-6 C atoms, the particles of the storage alloy being covered with PTFE in the manner of fibrils.

The addition of soot is important to make the mixture easier to process. The addition of soot makes the mixture pasty and able to flow. In the electrode, the soot promotes the electrical contact on a microscale (up to approximately 500 μm), i.e. it spans the distances and provides electrical contact between the openings or pores in the substrate material. These distances or openings, with a size of up to 500 μm , cannot be bridged by the particles in the paste or the active compound which can be obtained therefrom, since they generally have a diameter of only approximately 10 to 100 μm . Furthermore, the soot serves as an oxygen getter for protecting the oxygen-sensitive storage alloy.

The PTFE is responsible for the hydrophobic properties of the electrode and enables the three-phase boundary to be set. The reduction of the oxygen and the release and uptake of the hydrogen in the working cell take place in the only partially wetted pores. The PTFE is also responsible for enabling the paste to flow and hold together in the mixing or shaping process. The alcohol is in turn responsible for the temporary wetting of the PTFE powder, since otherwise there would be no distribution through fibrillation in the mixing process.

The method for producing the electrode according to the invention is very simple. The components are mixed in a mixer until a cohesive paste is formed. The paste is shaped and combined with the metallic substrate material (for example expanded metal, fabric, grid, perforated sheet) of the electrode. This is an extraordinarily simple process sequence.

The electrode according to the invention means that it is also no longer necessary to use a PTFE dispersion with a high wetting agent content, which has to be removed by decomposition at elevated temperature (300°C), damaging the storage alloy, in conventional plastic-bonded storage electrodes.

The dry fraction contains 85-95 parts of the storage alloy,, approximately 2-10 parts of soot and 3-8 parts of PTFE. The liquid fraction contains 30-70 parts by volume of water and approximately 70-30 parts by volume of the alcohol. Alcohols with a boiling point of the order of magnitude of approximately 100°C, i.e. for example n-butanol or n-propanol, are particularly suitable.

Furthermore, polyethylene glycol (PEG) may be included in the liquid fraction. The PTFE component means that the finished electrode can only be wetted by lye with extreme difficulty. Therefore, to achieve a sufficient uptake of electrolyte, a

polyethylene glycol can be supplied with the make-up water. The proportion of polyethylene glycol is 0.05-0.2% (based on the dry fraction). It is preferable to use a polyethylene glycol with a molecular weight of between 10^5 and 5×10^6 g/mol. The alcohol used is preferably n-propanol or n-butanol. Depending on the soot content, the ratio of the dry fraction to the liquid fraction is between 4:1 and 6:1, based on mass.

The electrode according to the invention is preferably used in an alkaline storage battery with positive nickel oxide electrode.

The electrode according to the invention is produced by rolling a dough-like paste onto a structured metal substrate, such as for example an expanded metal or grid mesh. The dough-like paste is prefabricated in a first production step by a mixing and kneading process. The solid and liquid components are mixed in a kneading machine until a cohesive paste is formed, for example in a stable domestic kneading machine. The PTFE particles are fibrillated by the hard compound grains and hold the paste together. The electrode is shaped either by manual rolling or in a rolling train. Either a sheet is produced and is combined with the substrate after drying or the kneaded compound is applied directly to the substrate and is then dried.

We claim:

1. A gastight cell for the electrochemical storage of energy, having at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and having an alkaline electrolyte or an alkaline electrolyte mixture, characterized in that one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.
2. The cell as claimed in claim 1, characterized in that the outer negative electrode of an electrode assembly comprising n positive electrodes and $(n+1)$ negative electrodes are flanked by in each case one transport element.
3. The cell as claimed in claim 1, characterized in that one or more negative electrodes are split into two parts, the two parts being separated from one another by a hydrophobic, gas-permeable transport element.
4. The cell as claimed in claim 3, characterized in that in the sequence of a plurality of positive and negative electrodes, every second negative electrode is split into two parts.
5. The cell as claimed in one of claims 3 to 4, characterized in that the two parts of the split negative electrodes have half the thickness or half the capacitance of the unsplit negative electrodes.
6. The cell as claimed in one of the preceding claims, characterized in that the transport element is a

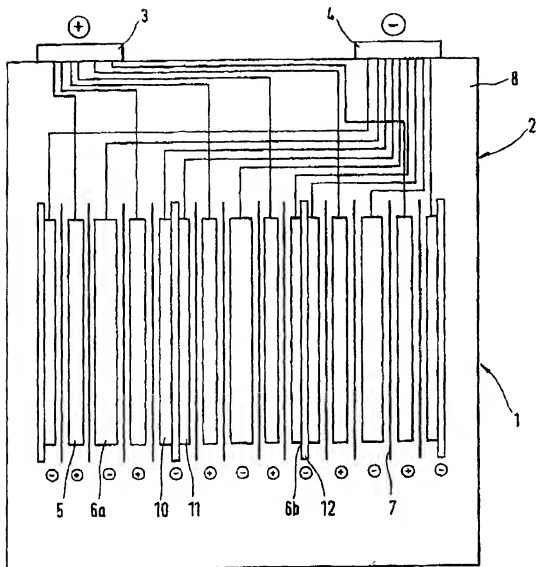
hydrophobic nonwoven layer, preferably comprising electrolyte-repelling polypropylene fibers.

7. The cell as claimed in one of the preceding claims, characterized in that the positive electrodes are fibrous-structure framework electrodes.
8. The cell as claimed in one of the preceding claims, characterized in that the separators comprise polyamide fiber nonwoven or hydrophilic polypropylene fiber nonwoven.
9. The cell as claimed in one of the preceding claims, characterized in that it has an electrode with a capacity for storing hydrogen, having a metallic substrate material, to which an active compound is applied, the active compound being obtainable from a paste which comprises a dry fraction and a liquid fraction, the dry fraction consisting of a mixture of a pulverulent storage alloy for hydrogen, soot and polytetrafluoroethylene (PTFE), the particles of the storage alloy being covered with PTFE in the manner of fibrils, and the liquid fraction consisting of a mixture of water and an alcohol which has 3 to 6 C atoms.
10. The cell as claimed in claim 9, characterized in that the dry fraction contains 85 to 95 parts of the alloy for storing hydrogen, 2 to 10 parts of soot and 3 to 8 parts of PTFE.
11. The cell as claimed in one of claims 9 to 10, characterized in that the liquid fraction contains 30 to 70 parts by volume of water and 70 to 30 parts by volume of the alcohol, as well as 0.05 to 0.2% (based on the dry fraction) of PEG.

12. The cell as claimed in one of claims 9 to 11, characterized in that polyethylene glycol (PEG) is also included in the liquid fraction.
13. The cell as claimed in one of claims 9 to 12, characterized in that the mass ratio of the dry fraction to the liquid fraction is 4:1 to 6:1.

Abstract

The present invention relates to a gastight nickel/metal hydride cell for the storage of electrochemical energy, having at least one positive nickel oxide electrode and at least one hydrogen-storing negative electrode, a hydrophilic separator being arranged between the positive and negative electrodes, and an alkaline electrolyte or an alkaline electrolyte mixture, in that one or more negative electrodes are provided with a gas-permeable, hydrophobic transport element for transporting the gases of the cell atmosphere.



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COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (includes Reference to PCT International Applications)

ATTORNEYS DOCKET NUMBER

979/50806

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

GASTIGHT, PRISMATIC NICKEL/METAL HYDRIDE CELL

the specification of which (check only one item below):

- ☐ is attached hereto.
- ☐ was filed as United States application
Serial No. _____
on _____
And was amended
on _____ (if applicable).
- ☒ was filed as PCT international application
Number PCT/EP00/05377
on 10 June 2000
and was amended under PCT Article 19
on _____ (if applicable).

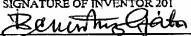
I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a).

I hereby claim foreign priority benefits under Title 35, United State Code, § 119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:

COUNTRY (if PCT indicate PCT)	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 119
Germany	199 29 947.1	29 June 1999	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No

Combined Declaration For Patent Application and Power of Attorney (Continued) (includes Reference to PCT international Applications)				ATTORNEY'S DOCKET NUMBER 979/50806	
I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national of PCT international filing date of this application.					
PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. 120					
U.S. APPLICATION NUMBER		U.S. APPLICATIONS U.S. FILING DATE		STATUS (Check one) PATENTED PENDING ABANDONED	
PCT APPLICATIONS DESIGNATING THE U.S.					
PCT APPLICATION NO	PCT FILING DATE	U.S. SERIAL NUMBERS ASSIGNED (IF ANY)			
POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (List name and registration number)					
Herbert I. Cantor, Reg. No. <u>24,392</u> ; James F. McKeown, Reg. No. <u>25,406</u> ; Donald D. Evenson, Reg. No. <u>26,160</u> ; Joseph D. Evans, Reg. No. <u>26,269</u> ; Gary R. Edwards, Reg. No. <u>31,824</u> ; and Jeffrey D. Sanok, Reg. No. <u>32,169</u>					
Send Correspondence to: Crowell & Moring, L.L.P., P.O. Box 14300 Washington, D.C. 20044-4300				Direct Telephone Calls to. (name and telephone number) (202) 624-2500	
100 201	FULL NAME OF INVENTOR	FAMILY NAME <u>BENCZUR LERMOESSY</u>	FIRST GIVEN NAME <u>Gabor</u>	SECOND GIVEN NAME	
	RESIDENCE & CITIZENSHIP	CITY <u>Stuttgart</u>	STATE OR FOREIGN COUNTRY <u>Germany</u>	COUNTRY OF CITIZENSHIP <u>Germany</u>	
	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>Nachtgallenweg 10 b</u>		STATE & ZIP CODE/COUNTRY <u>D-70199 Germany</u>	
202	FULL NAME OF INVENTOR	FAMILY NAME	FIRST GIVEN NAME	SECOND GIVEN NAME	
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP	
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE & ZIP CODE/COUNTRY	
203	FULL NAME OF INVENTOR	FAMILY NAME	FIRST GIVEN NAME	SECOND GIVEN NAME	
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP	
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SIGNATURE OF INVENTOR 201 		SIGNATURE OF INVENTOR 202		SIGNATURE OF INVENTOR 203	
DATE <u>January 13, 2002</u>		Date		DATE	